

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
3 January 2002 (03.01.2002)

PCT

(10) International Publication Number
WO 02/00793 A1

- (51) International Patent Classification⁷: **C08L 77/06**, 69/00
- (21) International Application Number: **PCT/NZ01/00124**
- (22) International Filing Date: **26 June 2001 (26.06.2001)**
- (25) Filing Language: **English**
- (26) Publication Language: **English**
- (30) Priority Data:
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- (81) Designated States (*national*): AE, AG, AL, AM, AT, AT
(utility model), AU, AZ, BA, BB, BG, BR, BY, BZ, CA,
CH, CN, CO, CR, CU, CZ, CZ (utility model), DE, DE
(utility model), DK, DK (utility model), DM, DZ, EC, EE,
EE (utility model), ES, FI, FI (utility model), GB, GD, GE,
GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ,
LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN,
MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI,
SK, SK (utility model), SL, TJ, TM, TR, TT, TZ, UA, UG,
US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (*regional*): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian
patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European
patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE,
IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF,
CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
- Published:**
— *with international search report*
- For two-letter codes and other abbreviations, refer to the "Guid-
ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.*



WO 02/00793 A1

(54) Title: A MODIFIED POLYAMIDE AND POLYCARBONATE BLEND

(57) Abstract: A polyamide/polycarbonate blend including Nylon 66, a polycarbonate and a compatibilizer selected from a maleic anhydride or epoxy modified amorphous polymer.

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A MODIFIED POLYAMIDE AND POLYCARBONATE BLEND

FIELD OF INVENTION

5 This invention involves a modified blend of polyamide, specifically Nylon-66 (polyhexamethylene adipamide), and polycarbonate which has improved toughness and is more easily processed than conventional unmodified blends.

10 BACKGROUND TO THE INVENTION

 Polyamide/polycarbonate blends have been of interest to both scientists and industry because of their anticipated superior properties to the separate constituent homopolymers. Nylon-6/polycarbonate
15 blends have been successfully developed, but it has proved much more difficult to produce commercially useful Nylon-66/polycarbonate blends.

 There has been considerable investigation of polymer blends,
20 but generally on a fairly ad hoc basis without any particular regard to the microstructure and the relationship between the structure and chemical and physical properties. In most case, polymer blends are incompatible, in that the two or three components do not mix well. A compatibilizer is needed, but considerable research is required to
25 identify what kind of compatibilizer is appropriate and, to date, the preferred compatibilizer and blend of components necessary to produce a commercially useful Nylon-66/polycarbonate blend have not been identified.

US 5,478,887 discloses a polyamide/polycarbonate blend employing the use of an elastomeric compatibilizer, which is claimed to have improved strength and high flexural modulus. The blends disclosed all involve polyamides in the form of Nylon-6, Nylon-12 or
5 Nylon-12,12. The physical properties of the Nylon-6, Nylon-12 and Nylon-12,12 polyamides result in a blend having physical characteristics, and therefore potential commercial application, different from those which would be anticipated for a Nylon-66/polycarbonate blend. Furthermore, the different chemical
10 properties of the polyamides of these known blends from those of Nylon-66 do not permit extrapolation from one to the other.

Since Nylon-66 has some advantages over Nylon-6, such as its electrical properties, it would be beneficial to develop commercially
15 useful blends of Nylon-66 and polycarbonate. Such a blend may be preferred for use in electrical fittings, motor vehicle parts, computer and related technology and certain household and office equipment, for example.

20 **OBJECT OF THE INVENTION**

With the above discussion in mind, it is an object of the present invention to provide a polyamide/polycarbonate blend which reduces or overcomes at least some of the above-mentioned problems, or which at least provides the public with a useful alternative.

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Other objects of the invention may become apparent from the following description which is given by way of example only.

SUMMARY OF THE INVENTION

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According to one aspect of the present invention there is provided a polyamide/polycarbonate blend including:

- Nylon 66 in a proportion of substantially between 10 and 90 parts by weight,
- polycarbonate in a proportion of substantially between 90 and 10 parts by weight,
- a compatibilizer selected from a maleic anhydride or epoxy modified amorphous polymer, in the proportion of substantially between 2 and 30 parts by weight, and
- a toughener, selected from rubbers or rubber-toughened amorphous polymers, in the proportion of 0 to 40 parts by weight.

In one preferred form the compatibilizer may be maleic anhydride-grafted styrene/ethylene-butylene/styrene copolymer (SEBS-g- MA).

Preferably, the compatibilizer may be included in the proportion substantially 5 to 10 parts by weight.

In a further preferred form the Nylon 66 may be included in a proportion of substantially 10 to 30 parts by weight and the polycarbonate in the proportion of substantially 90 to 70 parts by weight.

In an alternative preferred form the Nylon 66 may be included in a proportion of substantially 70 to 90 parts by weight and the polycarbonate in a proportion of substantially 30 to 10 parts by weight.

In one preferred form the blend may preferably include tougheners in the proportion of substantially 5 to 15 parts by weight.

Preferably the tougheners may be selected from styrene/ethylene-butylene-/styrene copolymer (SEBS), Paraloid EX-3361, acrylonitrile/butylene/styrene copolymer and high-impact polystyrene. More preferably the toughener may be styrene/ethylene/butylene/styrene.

In one preferred form the blend includes Nylon 66 substantially 70 parts by weight, polycarbonate substantially 20 parts by weight SEBS substantially 5 parts by weight and SEBS-g-MA substantially 5 parts by weight.

According to a further aspect of the present invention there is provided a polycarbonate/polyamide melt compound blend in which the polyamide is Nylon 66 and which includes a proportion of a compatibilizer selected from a maleic anhydride and epoxide modified amorphous polymers in a sufficient proportion to produce a blend which is processable and has a commercially useful impact strength.

According to further aspect of the present invention there is provided a product formed from a polyamide/polycarbonate blend as will be herein described.

According to a further embodiment, there is provided a method for producing a polyamide/polycarbonate blend including the step of melt blending Nylon 66, a polycarbonate and a compatibilizer selected from a maleic anhydride or epoxy modified amorphous polymer in the proportions of Nylon 66 10-90 parts by weight, polycarbonate 90-10 parts by weight; compatibilizer 2-30 parts by weight.

Preferably, the blending temperature is between about 255°C and 275°C.

Preferably the method further includes the steps of extruding the blend followed by pelletising.

Preferably the method further includes the step of injection moulding to form a product.

Preferably injection moulding occurs at temperatures between about 245°C and 280°C.

According to a further aspect of the present invention there is provided a polyamide/polycarbonate blend substantially as herein described and with reference to the accompanying figures.

BRIEF DESCRIPTION OF THE FIGURES

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Figure 1: Relationship between toughener concentration, compatibilizer concentration and toughness of PA66/PC blend with 70 parts by weight of PA66 and 20 parts by weight of PC.

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Figure 2: The effect of compatibilizer and on tensile strength and elongation of PA66/PC blends.

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Figure 3: The effect of compatibilizer concentration and toughener on tensile strength and elongation of PA66/PC blends.

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Figure 4: Effect of compatibilizer (SEBS-g-MA) and different toughener on the morphology of the PA66/PC blend: A. toughener (SEBS) only; B. toughener + compatibilizer; C. toughener (HIPS) only; D. toughener + compatibilizer; E. toughener (Paraloid EX3361) only; F. toughener +

compatibilizer; G. toughener (SEBS) only; H.
toughener + compatibilizer.

Figure 5: Effect of compatibilizer (SEBS-g-MA)
concentration on the morphology of the PA66/PC
blend.

Figure 6: Effect of compatibilizer (SEBS-g-MA) and
toughener (SEBS) concentration on the
morphology of the PA66/PC blend.

Figure 7: Effect of blend composition on flexural strength.
Sample composition is shown in Table 5.

DETAILED DESCRIPTION OF THE INVENTION

In broad terms this invention involves the blending of the
polyamide Nylon-66 with polycarbonate using a compatibilizer selected
to improve toughness and for its ability to remain in the interphase
after it reacts with the polyamide. The compatibilizer reacts with the
polyamide Nylon-66 and also mixes well, but not completely, with
polycarbonates. The properties of the blend can be tailored by
adjusting the composition of the blend which could include the addition
of a toughening agent.

The polyamide Nylon 66 is well known. It may be produced by
condensation polymerisation of hexamethylene tetramine and adipic
acid, or by condensation polymerisation of the 1:1 salt formed by
hexamethylene tetramine and adipic acid, as will be well known to the
skilled person.

The specific polycarbonate employed in the blends of the
invention is not important, as will be appreciated by those skilled in

the art, but for practical and commercial reasons the selected polycarbonate used herein was bisphenol-A polycarbonate. Other polycarbonates that could be used include various compounds such as the polycarbonates listed in US 5008347 (the disclosure of which is
5 incorporated herein by reference) and many other patents. US 5008747 itself discloses a process for the production of polyalkyloxazoline-polycarbonate-polyalkyloxazoline triblock copolymers useful as a compatibilizing agent in the production of polyamide/polycarbonate resin blends.

10

The compatibilizer can be any maleic anhydride or epoxide modified amorphous polymer, the important characteristics being that the compatibilizer has reactive groups which can react with polyamides, and groups which are compatible with polycarbonate; an
15 example being maleic anhydride-grafted styrene/ethylene-butylene/styrene copolymer (SEBS-g-MA). The compatibilizer could, however, be any rubber or thermoplastic elastomer or amorphous plastic modified with reactive groups that will react with polyamides. Such compounds *per se* would be well known to a skilled person.

20

Additional tougheners which may be included can be rubbers or rubber-toughened amorphous polymers, such as poly(styrene-ethylene-butylene-styrene) (SEBS), high-impact polystyrene (HIPS), acrylonitrile-butylene-styrene copolymer (ABS) and Paraloid EX-3361. Other
25 tougheners that could be used include diene homopolymer rubbers, polychloroprene, polyisobutylene and butyl rubber, fluorine-containing rubbers, acrylic rubbers, rubbers produced by condensation and ring-opening polymerisation, polyurethane rubbers and thermoplastic rubbers.

30

The proportions of the components should be sufficient to provide a blend which is processable to form a product having a commercially useful impact strength.

The polyamide Nylon-66 should be included in the proportion of 10-90 parts by weight, but preferably 10-30 parts or 70-90 parts. The polycarbonate included in proportions of 90-10 parts by weight, preferably 90-70 parts by weight or 30-10 parts by weight. The compatibilizer included in the proportion 2-30 parts by weight of the whole blend, preferably 5-10 parts. Additional tougheners may be included in the proportion 0-40 parts by weight of the blend, preferably 5-15 parts. A preferred toughener is SEBS.

10

The polyamide/polycarbonate blends may be formed by conventional methods involving screw extruder, for example an Axon Model BX-18-286 single screw extruder. The process for producing the blend includes the step of melt blending the components at temperatures in the range of about 255-275°C in the extruder, followed by extrusion through a die as a single strand or multiple strands, water cooling the extruded strand or strands and pelletising.

Subsequent processing to produce a product can be carried out for example by injection moulding at 245-280°C in a machine such as a BOY model 50M automatic injection moulding machine.

The temperature selected for melt blending is dependent on the tougheners which are optionally included in the blend. For those tougheners susceptible to decomposition at higher temperatures (eg Paraloid EX3361), the process is carried out at temperatures in the lower region of the range, (eg 255-265°C) whilst for other blends temperatures at the higher end of the range are preferred.

Tensile strength, tensile modulus, flexural strength, flexural modulus and Charpy impact strength were measured according to the ASTM D638-98, D256-93a and D790-98 protocols.

Table 1 and Figure 1 show the general trend of an increase in toughness (as measured by impact strength) with increasing proportions of toughener and compatibilizer. There are some exceptions to the general trend: (a) Paraloid EX-3361 alone can improve the toughness of the blend; (b) incorporation of ABS and compatibilizer enhances processability of the blend, but ABS with or without compatibilizer does not improve the blend's toughness.

The polycarbonate ("PC") used was bisphenol-A carbonate.
PA66 refers to polyamide 66 (ie. Nylon 66).

Table 1. Effects of compatibilizer concentration and toughener concentration on toughness of PA66/PC blends.

Sample Number	Sample Composition(parts by weight)	Impact Strength (J/m)	Note *
1	PA66/PC (70/20)	23.4	Reference
2	PA66/PC/HIPS (70/20/10)	25.0	T
3	PA66/PC/ABS/SEBS-g-MA (70/20/5/5)	25.0	T + C
4	PA66/PC/ABS (70/20/10)	26.8	T
5	PA66/PC/SEBS (70/20/10)	27.4	T
6	PA66/PC/ Paraloid EX-3361 (70/20/10)	48.1	T
7	PA66/PC/Paraloid EX-3361/SEBS-g-MA (70/20/5/5)	51.4	T + C
8	PA66/PC/SEBS/SEBS-g-MA (70/20/5/5)	52.3	T + C
9	PA66/PC/SEBS-g-MA (70/20/10)	53.6	C
10	PA66/PC/HIPS/SEBS-g-MA (70/20/5/5)	57.0	T + C
11	PA66/PC/SEBS/SEBS-g-MA (70/20/10/10)	94.9	T + C
12	PA66/PC/SEBS-g-MA (70/20/20)	101	C
13	PA66/PC/SEBS-g-MA (70/20/30)	254	C
14	PA66/PC/SEBS/SEBS-g-MA (70/20/15/15)	280	T + C

* Note: T = Toughener; C = Compatibilizer.

Tensile strength and elongation are affected by addition of these modifiers (the tougheners and compatibilizer). For most of the blends, the addition of compatibilizer and tougheners increases

elongation at yield while decreasing tensile strength and modulus of elasticity. The magnitude of the changes in properties depends on the amount of additives (see Table 2 and Figures 2 and 3).

- 5 Table 2. The effect of compatibilizer and tougheners and their concentrations on the mechanical properties of PA66/PC blend.

Sample Composition (parts by weight)	Tensile Strength(MPa)	Elongation (%)	Modulus of Elasticity(MPa)
PA66/PC/SEBS-g-MA (70/20/10)	49.9	6.4	902
PA66/PC/SEBS-g-MA (70/20/20)	41.8	18.0	677
PA66/PC/SEBS-g-MA (70/20/30)	35.7	30.0	537
PA66/PC/SEBS/SEBS-g-MA (70/20/5/5)	50.0	4.8	906
PA66/PC/SEBS/SEBS-g-MA (70/20/10/10)	41.0	20.0	682
PA66/PC/SEBS/SEBS-g-MA (70/20/15/15)	34.8	26.4	534
PA66/PC/SEBS/(70/20/10)	57.3	3.3	1017
PA66/PC/Paraloid/SEBS-g-MA (70/20/5/5)	52.3	5.7	901
PA66/PC/Paraloid/(70/20/10)	56.6	3.8	959
PA66/PC/ABS/SEBS-g-MA (70/20/5/5)	36.5	4.5	724
PA66/PC/ABS (70/20/10)	56.7	2.2	1089
PA66/PC/HIPS/SEBS-g-MA (70/20/5/5)	49.4	11.0	880
PA66/PC/HIPS (70/20/10)	56.5	3.3	992

- 10 An increase in compatibilizer content, or both toughener (particularly SEBS) and compatibilizer content, produces a substantial increase in impact strength and elongation, and a reduction in tensile strength (see Figures 2 and 3).

- 15 A blend, in parts by weight, with preferred characteristics of strength and elasticity was 70 parts Nylon-66, 20 parts polycarbonate, 5 parts SEBS and 5 parts SEBS-g-MA.

The ratio of Nylon-66 to polycarbonate plays an important role in the mechanical properties (see Table 3). The preferred matrix of the blends should be either Nylon-66 or polycarbonate, so that the proportion of Nylon-66 should be 70-90 parts by weight or 10-30 parts by weight.

Table 3. The effect of Nylon-66 concentration on the toughness of the blends.

Sample Composition (part)	PA66/PC /SEBS (20/70/10)	PA66/PC/SE BS (50/50/10)	PA66/PC/ SEBS (70/20/10)	PA66/PC/ SEBS-g- MA (20/70/10)	PA66/PC/ SEBS-g- MA (50/50/10)	PA66/PC/ SEBS-g- MA (70/20/10)
Impact Strength (J/m)	25.0	19.6	27.4	66.2	25.9	53.6

Flexural properties of the blends are also affected by the addition of compatibilizers and tougheners. As for the tensile strength, the flexural strength and flexural modulus decrease as the concentration of compatibilizer and toughener increases.

The samples were prepared and tested according to the ASTM D790-98. The conditions are listed in Table 4. The results are given in Table 5 and Figure 7.

Table 4. Specimen preparation and test conditions.

Specimen preparation	Samples were injection moulded at 245 to 280°C with dimension: 126*12.47*3.08 and then kept in a desiccator for more than 48 hours before testing.
Procedure used (four points bending)	Procedure A and B, strain rate = 0.001 or 0.1 mm/mm/min.
Support span length	49.28 mm (L = 16d = 16*3.08 mm).
Rate of crosshead motion	11 mm/min.
Number of specimens tested	5 (minimum).
Maximum mid-span deflection	6.6 mm
Reason for rejecting specimens	Apparent flaws

5

Table 5. Effect of compatibilizer and toughener concentration on the flexural properties of the blends.

Sample ID	Sample Composition (parts)	Flexural Strength (MPa)	Modulus of Elasticity (MPa)
A	PA66/PC/ABS (70/20/10)	43.6	2636
B	PA66/PC/HIPS (70/20/10)	41.3	1759
C	PA66/PC/SEBS(70/20/10)	36.6	2106
D	PA66/PC/Paraloid(70/20/10)	42.2	2376
E	PA66/PC/SEBS-g-MA (70/20/10)	35.2	1856
F	PA66/PC/SEBS-g-MA (70/20/20)	31.8	1354
G	PA66/PC/SEBS-g-MA (70/20/30)	26.0	1017

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In order to study the relationship between the miscibility of the blends and the addition of tougheners and compatibilizer, scanning electron microscopy (SEM) was used to observe the change of domain size in the blends resulting from incorporation of tougheners and compatibilizer. Figures 4, 5 and 6 indicate that the addition of SEBS-g-MA as compatibilizer can improve the compatibility of the Nylon-66/PC blends, as shown by a decrease in domain size. We conclude that the compatibilizer is an indispensable component in Nylon-66/PC blend for enhanced miscibility of the component homopolymers.

Nylon-66 has some advantages over Nylon-6, such as in relation to electrical properties, which would make it a preferred choice in a polyamide/polycarbonate blend for use in, for example, electrical fittings, motor vehicle parts, computer and related technology, and certain household and office equipment. The blends of the present invention, which include the use of a selected compatibilizer with or without additional tougheners, form a Nylon-66/polycarbonate blend which also has improved toughness and can be processed more easily by existing technologies than can corresponding unmodified blends.

Where in the foregoing description reference has been made to specific components or integers of the invention having known equivalents then such equivalents are herein incorporated as if individually set forth.

Although this invention has been described by way of example and with reference to possible embodiments thereof it is to be understood that modifications or improvements may be made thereto without departing from the scope or spirit of the invention as embodied in the appended claims.

WHAT WE CLAIM IS:

1. A polyamide/polycarbonate blend including:
 - Nylon 66 in a proportion of substantially between
5 10 and 90 parts by weight,
 - polycarbonate in a proportion of substantially
between 90 and 10 parts by weight,
 - 10 - a compatibilizer selected from a maleic anhydride
or epoxy modified amorphous polymer, in the
proportion of substantially between 2 and 30
parts by weight.
- 15 2. A blend according to claim 1, wherein the compatibilizer is
maleic anhydride-grafted styrene/ethylene-butylene/styrene
copolymer (SEBS-g- MA).
3. A blend according to claim 1 or claim 2, wherein the
20 compatibilizer is included in the proportion substantially 5 to 10
parts by weight.
4. A blend according to claim 1, 2 or 3, wherein Nylon 66 is
25 included in a proportion of substantially 10 to 30 parts by
weight and the polycarbonate in the proportion of substantially
90 to 70 parts by weight.
5. A blend according to claim 1, wherein Nylon 66 is included in a
30 proportion of substantially 70 to 90 parts by weight and the
polycarbonate in a proportion of substantially 30 to 10 parts by
weight.
6. A blend according to any one of the preceding claims wherein

the polycarbonate is bisphenol-A polycarbonate.

7. A blend according to any one of the preceding claims, further including at least one toughener selected from rubbers or rubber-toughened amorphous polymers, in a proportion of up to about 40 parts by weight.
8. A blend according to any one of the preceding claims, wherein the polymer blend includes at least one toughener in the proportion of substantially 5 to 15 parts by weight.
9. A blend according to claim 7 or 8, wherein the toughener is selected from styrene/ethylene-butylene-/styrene copolymer (SEBS), Paraloid EX-3361, acrylonitrile/butylene/styrene copolymer and high-impact polystyrene.
10. A blend according to claim 9, wherein the toughener is styrene/ethylene-butylene/styrene (SEBS).
11. A blend according to claim 10, wherein the polymer blend includes substantially 70 parts by weight Nylon 66, substantially 20 parts by weight polycarbonate, substantially 5 parts by weight SEBS and substantially 5 parts by weight SEBS g-MA.
12. A polyamide/polycarbonate melt compound blend in which the polyamide in the blend is Nylon 66 and which includes a proportion of a compatibilizer selected from a maleic anhydride and epoxide modified amorphous polymers in a sufficient proportion to produce a polymer blend which is processable and has a commercially useful impact strength.
13. A method of producing a polyamide/polycarbonate blend

including the steps of melt blending Nylon 66, a polycarbonate, and a compatibilizer selected from a maleic anhydride or epoxy modified amorphous polymer, substantially in the proportions of:

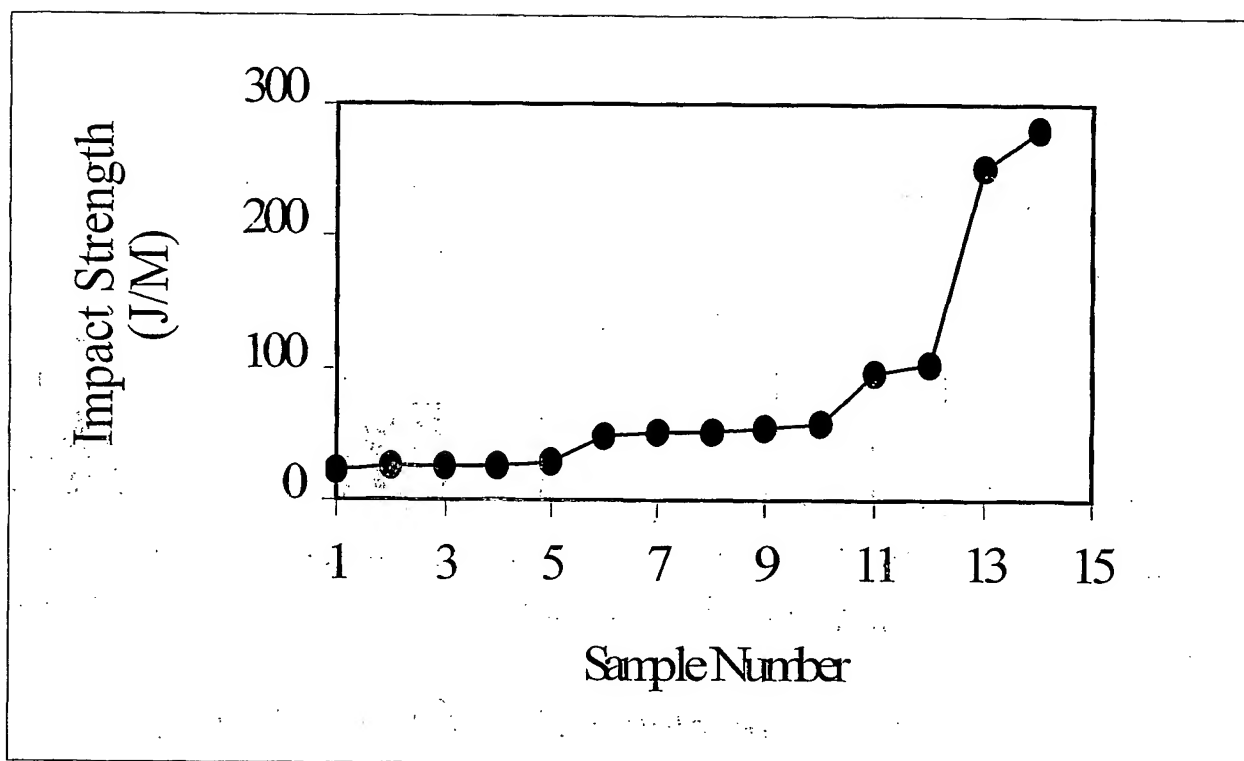
5

- (a) 10-90 parts by weight Nylon 66;
- (b) 90-10 parts by weight polycarbonate; and
- (c) 2-30 parts by weight compatibilizer.

- 10 14. The method according to claim 13 wherein a toughener is added to the melt blend in an amount of up to substantially 40 parts by weight.
- 15 15. The method according to claim 13 or 14 wherein the blending temperature is between about 235°C and 275°C.
16. The method according to any one of claims 13, 14 or 15 further including the steps of extruding the blend followed by pelletising.
- 20 17. The method according to claim 16 further including the step of injection moulding to form a product.
- 25 18. A polyamide/polycarbonate blend substantially as herein described and with reference to the accompanying Figures and Examples.

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**Figure 1**

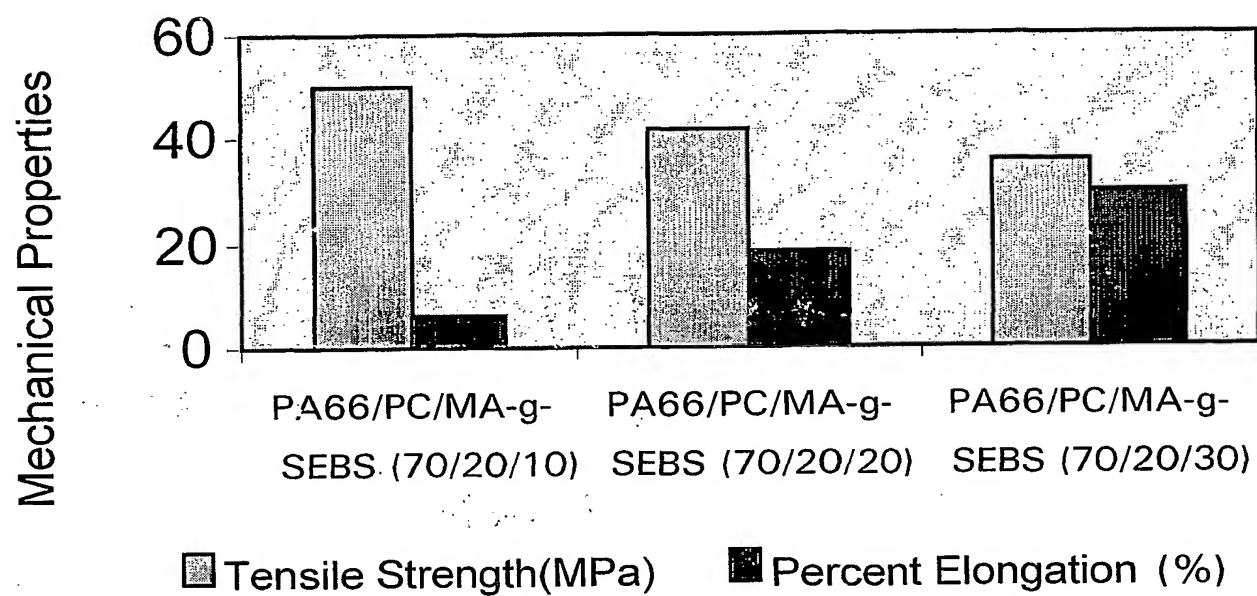


Figure 2

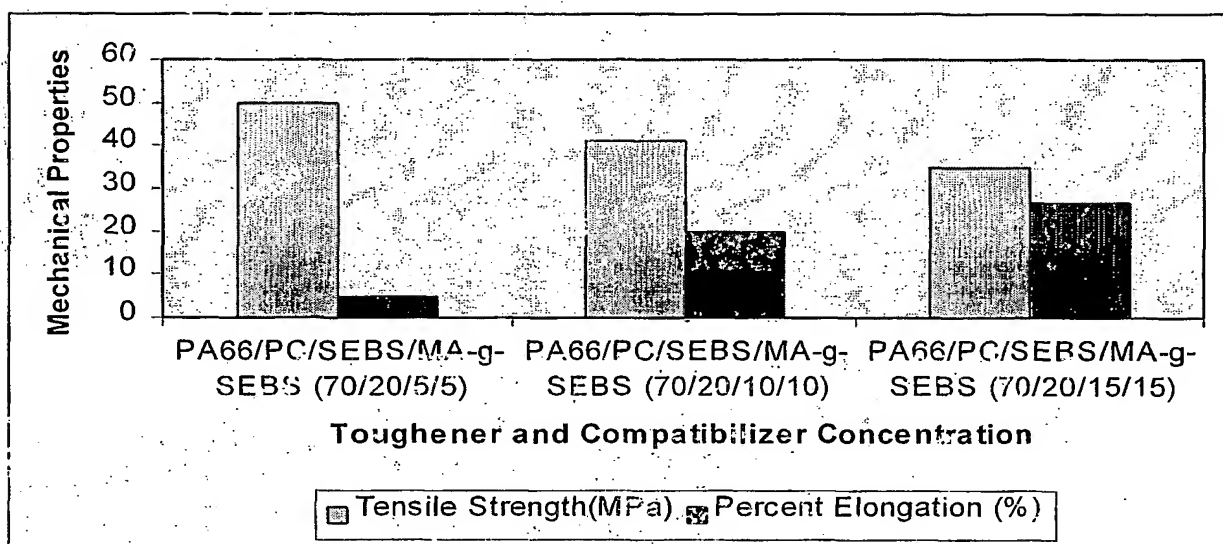
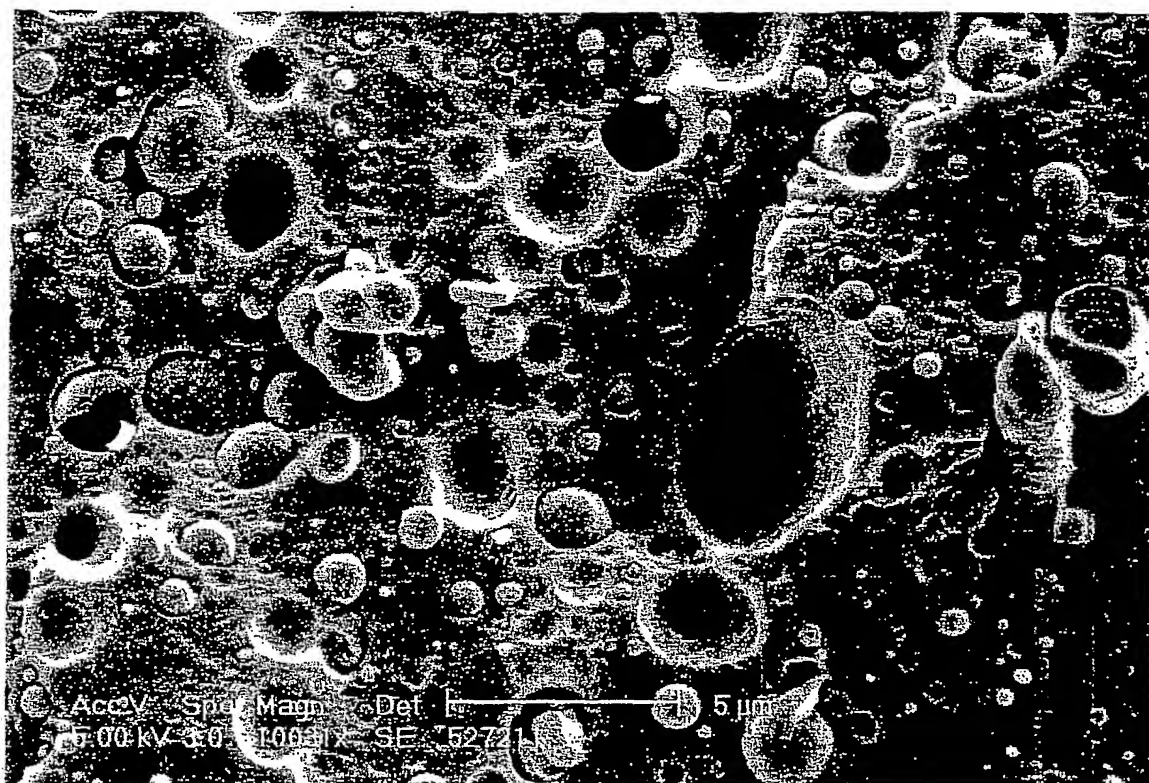
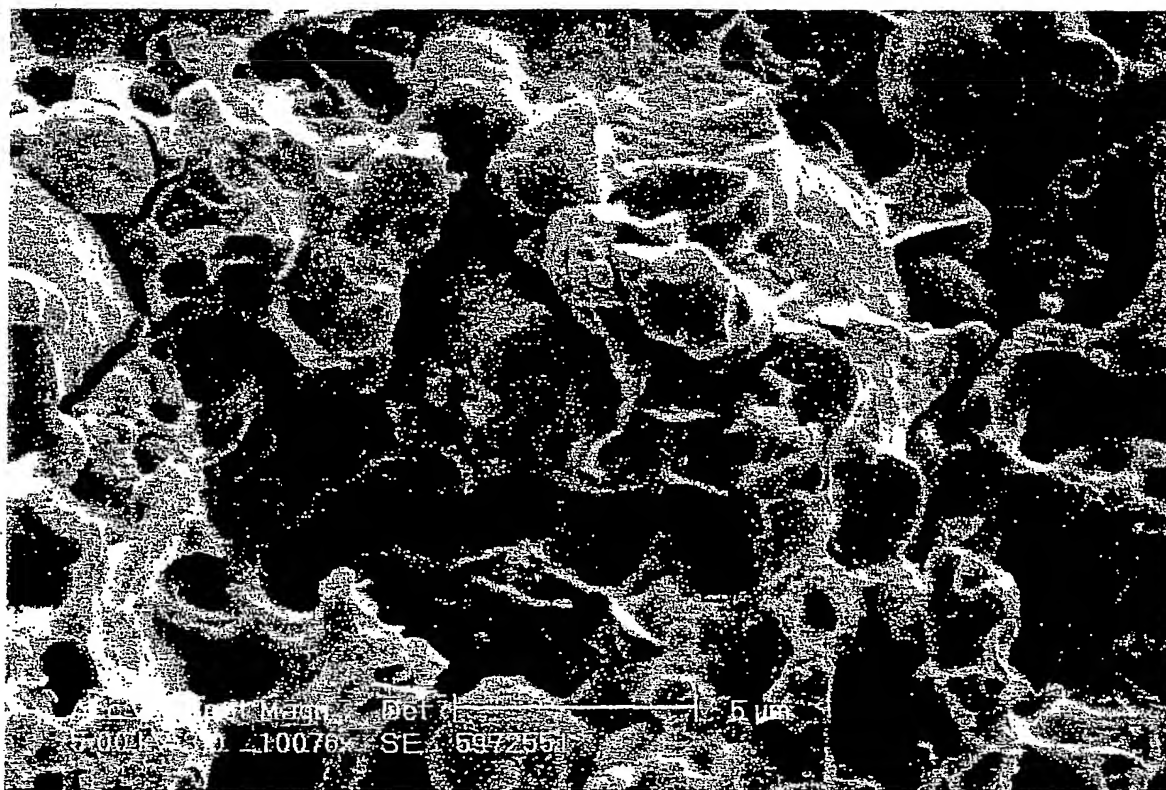


Figure 3



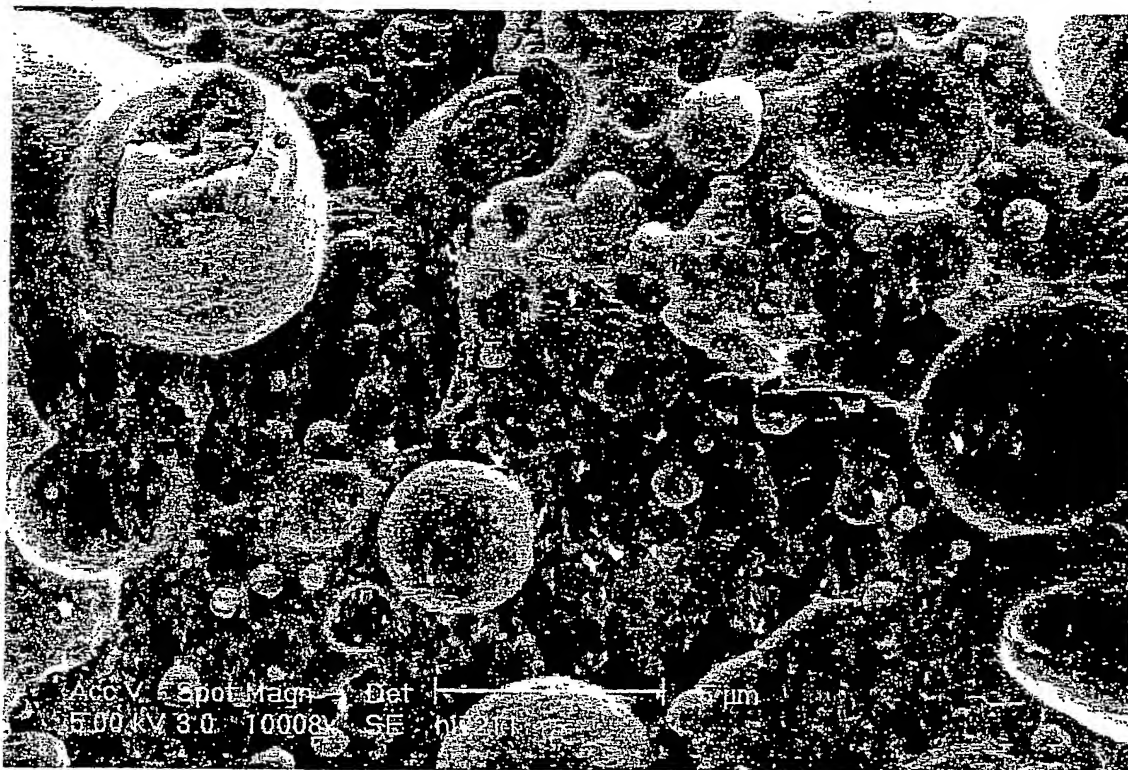
A. SEM of PA66/PC/SEBS Blend (70/20/10)



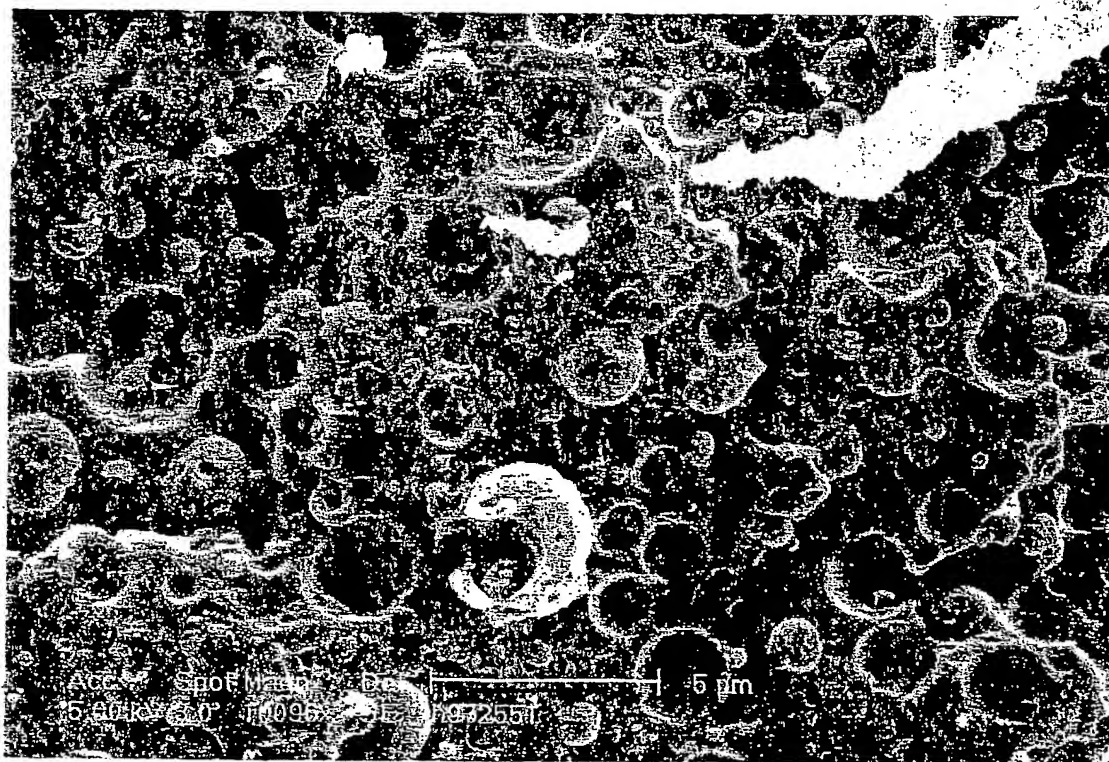
B. SEM of PA66/PC/SEBS/SEBS-g-MA Blend (70/20/5/5)

FIGURE 4

A and B



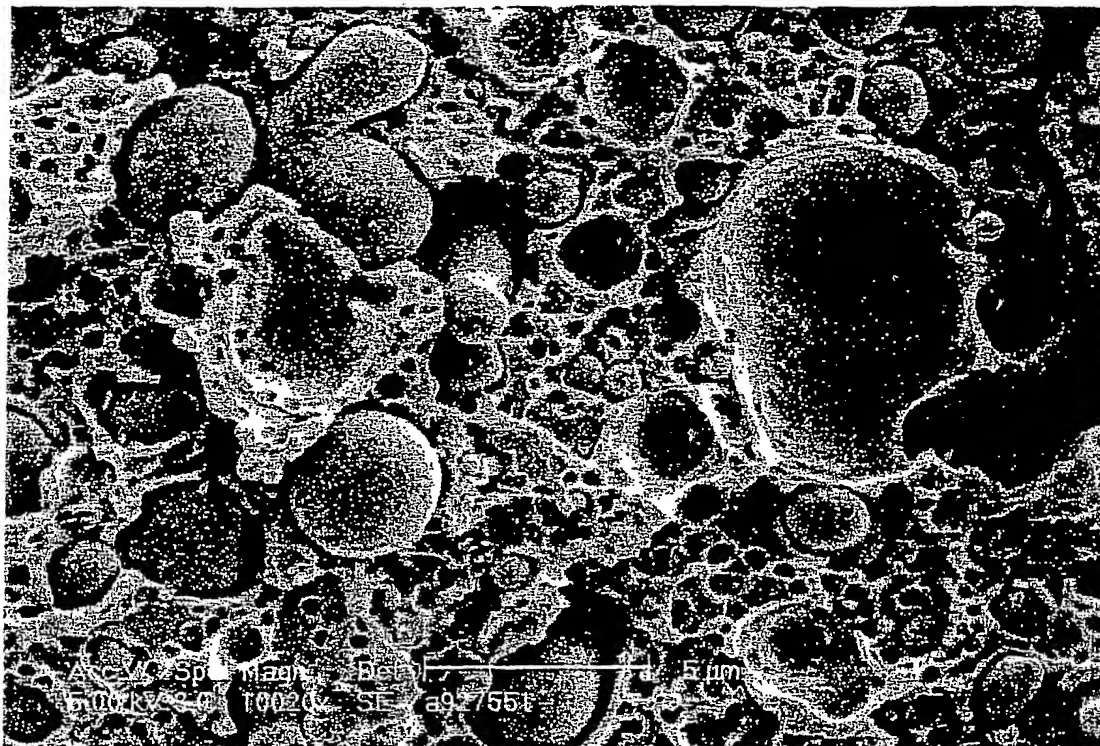
C. SEM of PA66/PC/HIPS Blend (70/20/10)



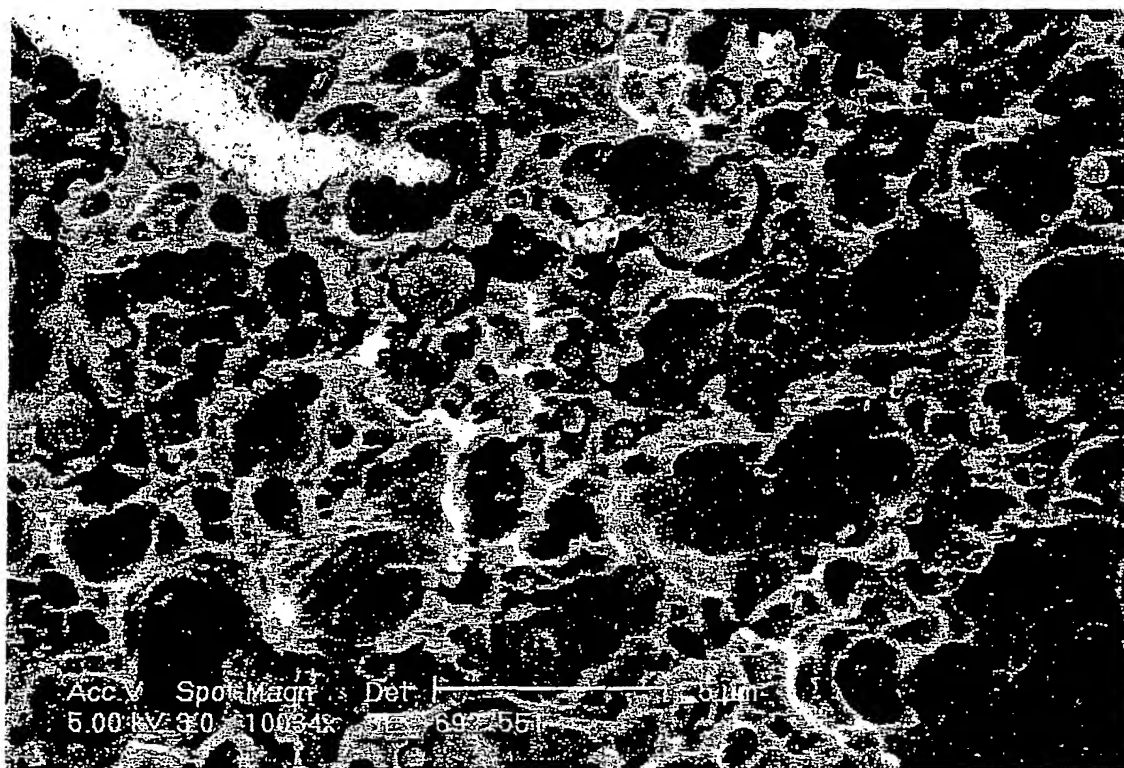
D. SEM of PA66/PC/HIPS/SEBS-g-MA Blend (70/20/5/5)

FIGURE 4

C and D

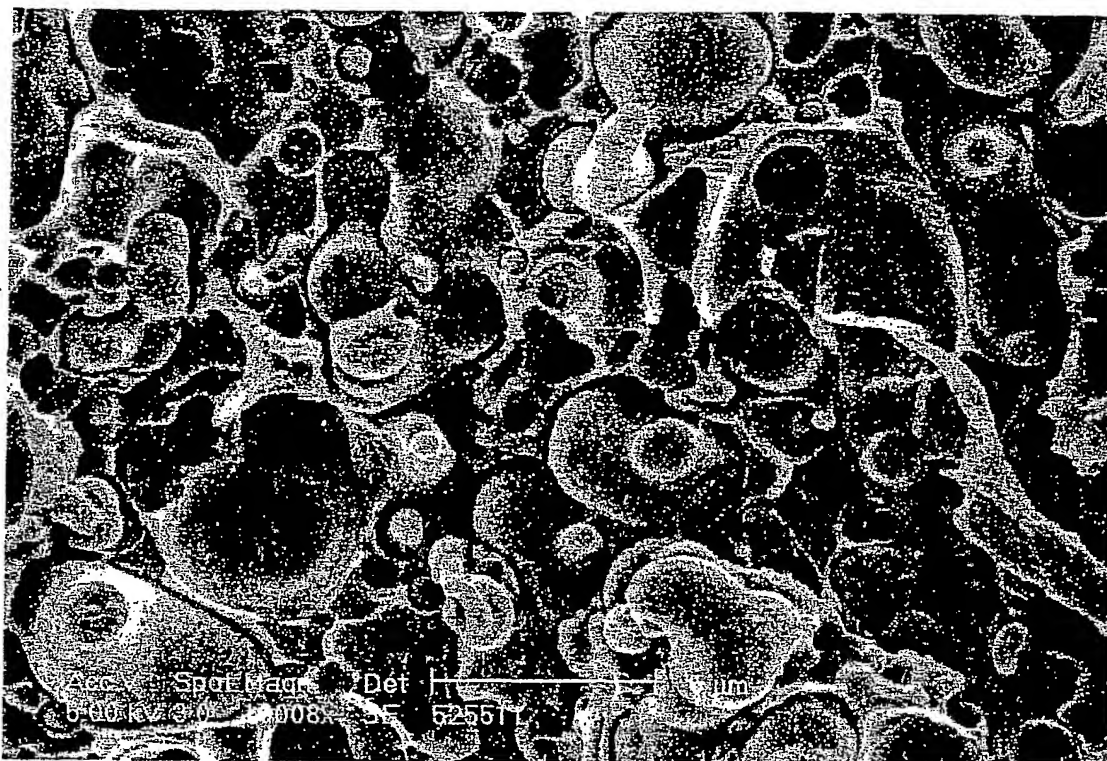


E. SEM of PA66/PC/Paraloid Blend (20/70/10)

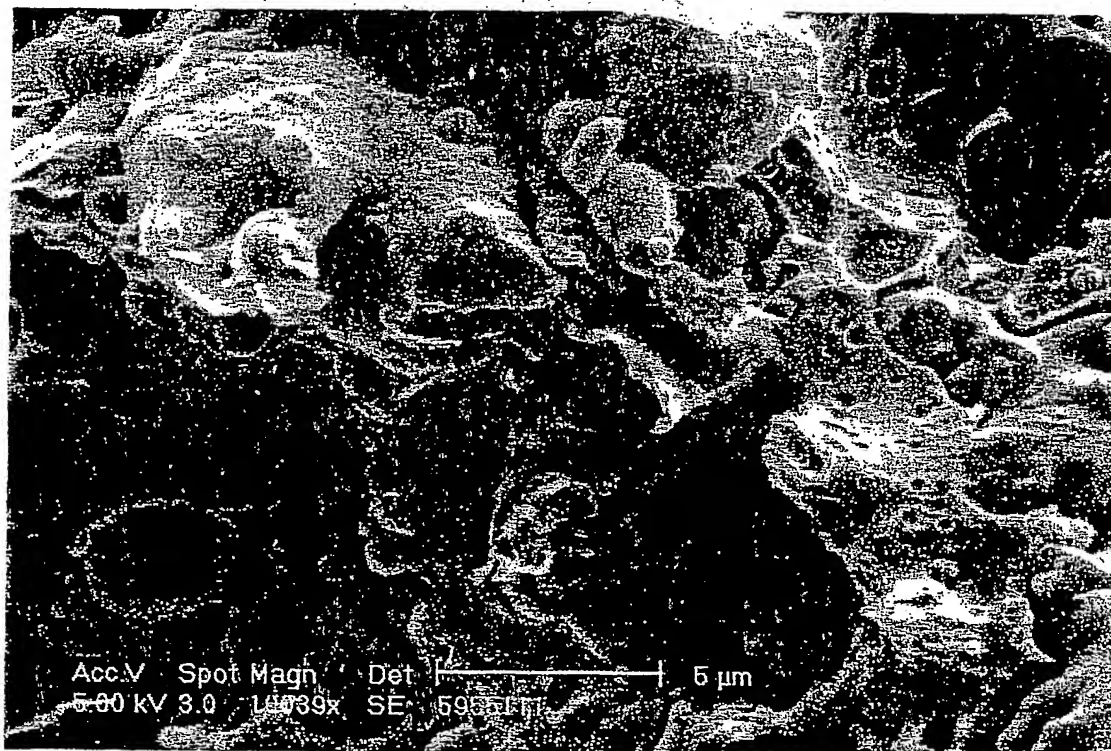


F. SEM of PA66/PC/Paraloid/SEBS-g-MA Blend (20/70/5/5)

FIGURE 4
E and F



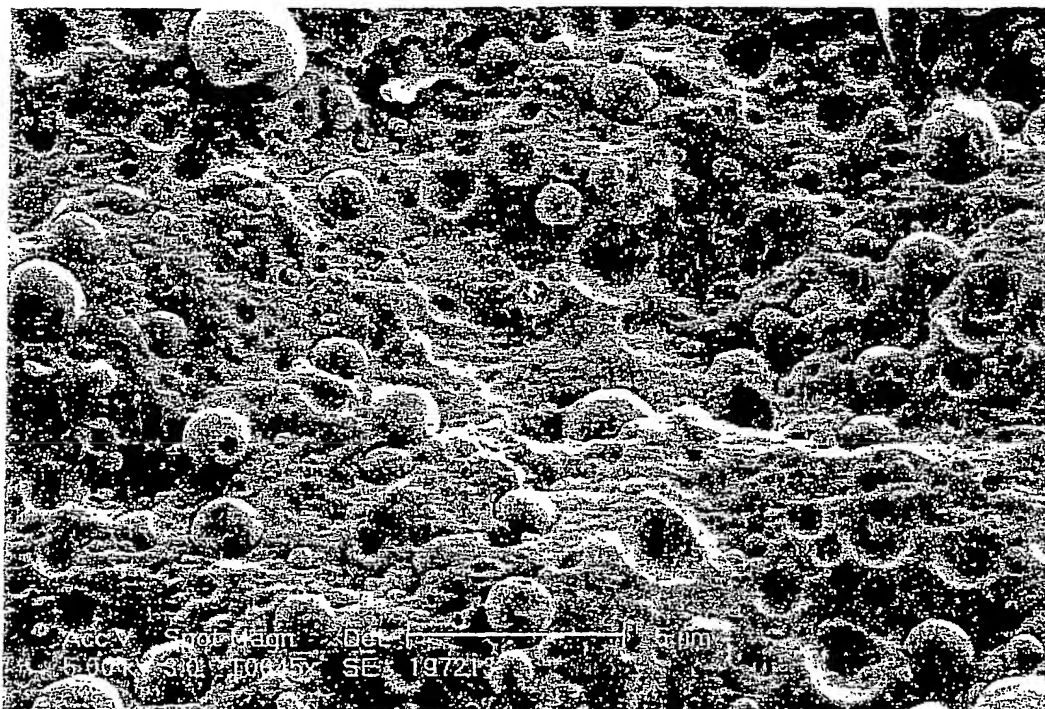
G. SEM of PA66/PC/ SEBS Blend (50/50/10)



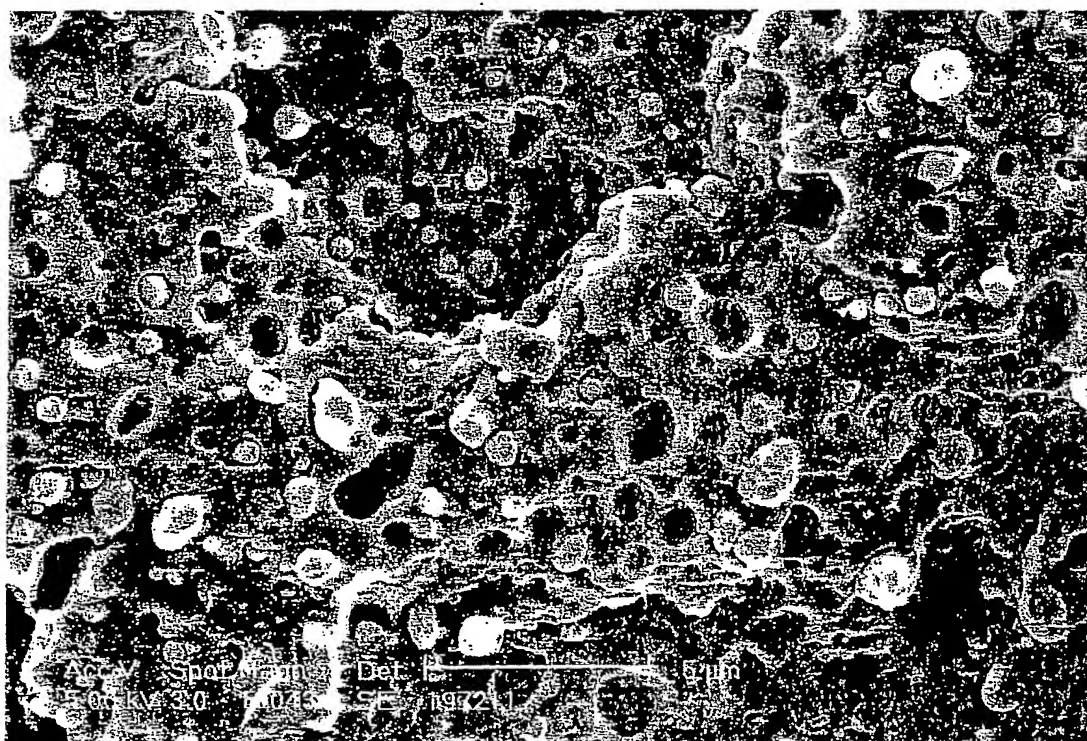
H. SEM of PA66/PC/ SEBS/SEBS-g-MA Blend (50/50/5/5)

FIGURE 4

G and H

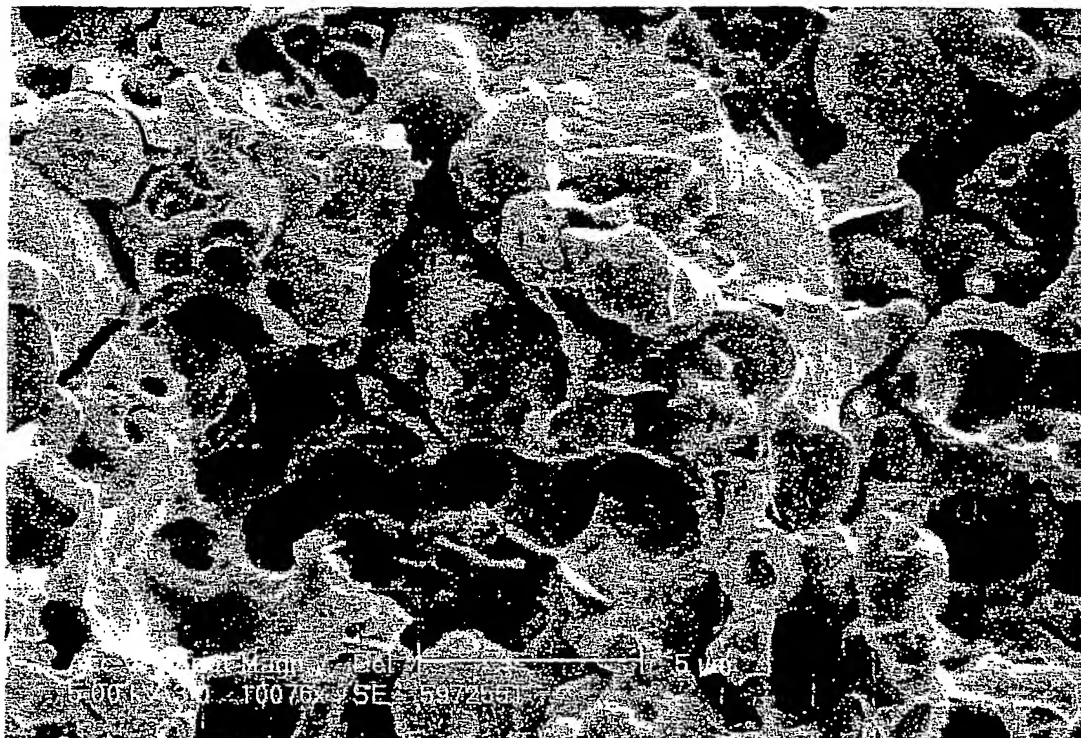


A. SEM of PA66/PC/ SEBS-g-MA Blend (70/20/10)

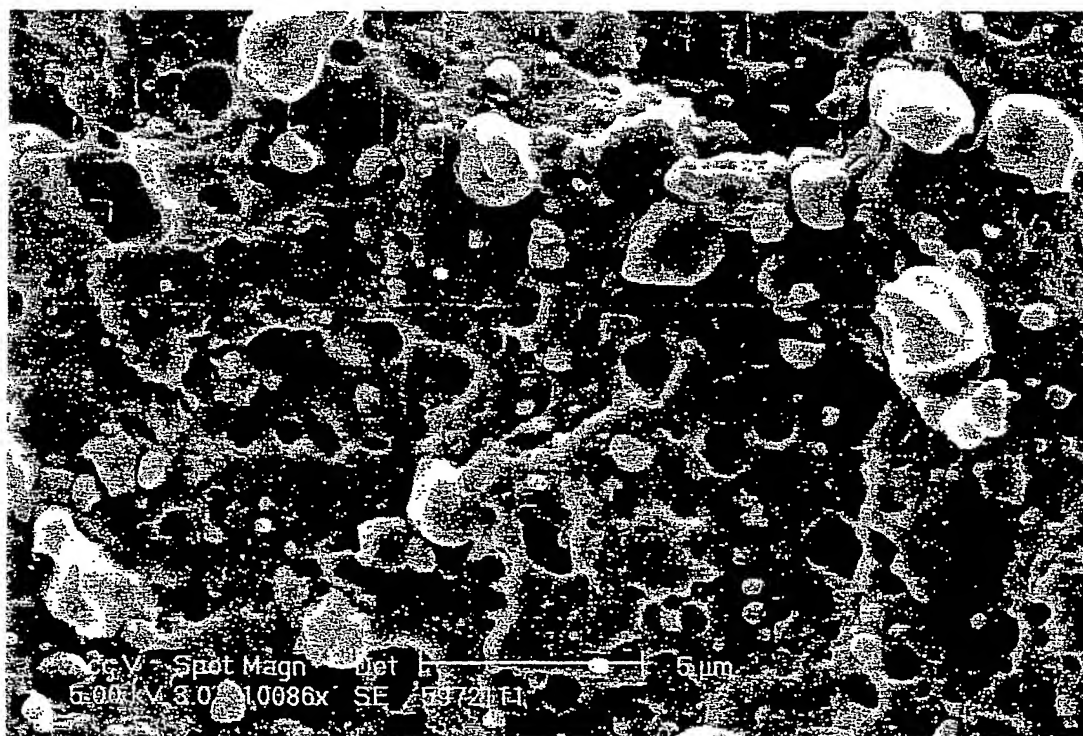


B. SEM of PA66/PC/ SEBS-g-MA Blend (70/20/20)

FIGURE 5



A. SEM of PA66/PC/SEBS/ SEBS-g-MA Blend (70/20/5/5)



B. SEM of PA66/PC/SEBS/ SEBS-g-MA Blend (70/20/10/10)

FIGURE 6

- 5 Effect of blend composition on flexural strength. Sample composition is shown in Table 5.

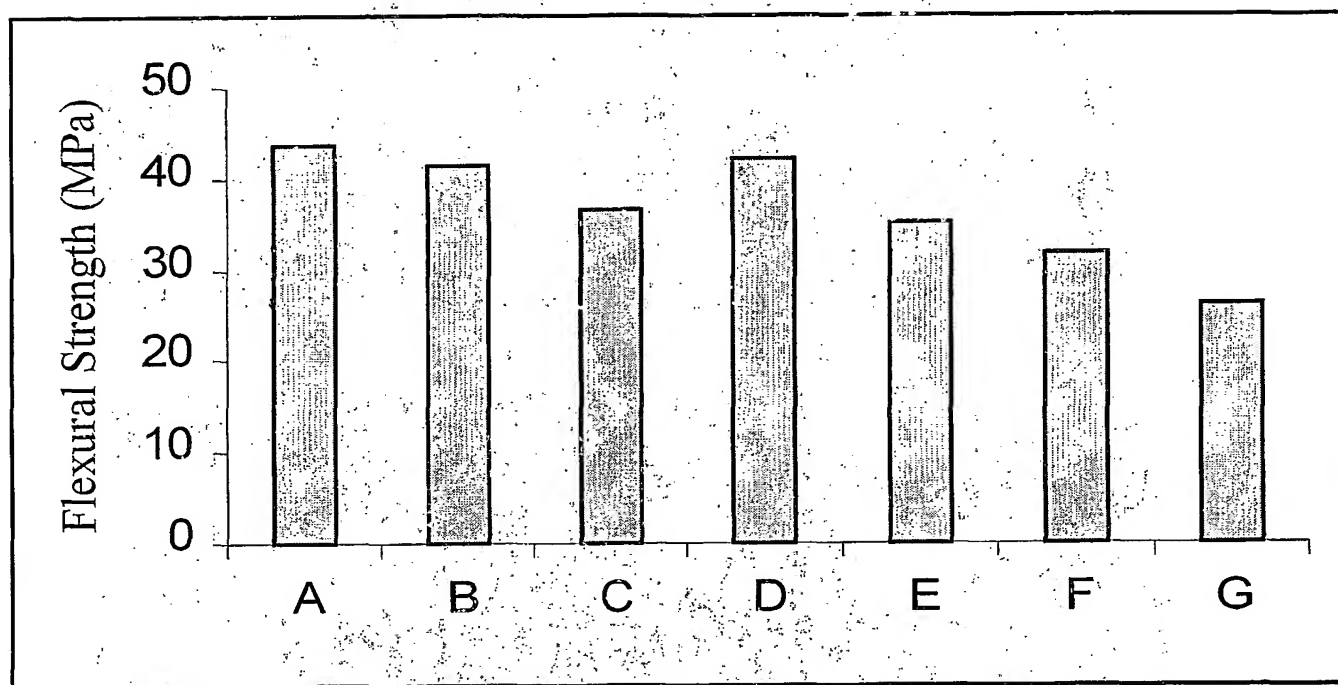


Figure 7

INTERNATIONAL SEARCH REPORT

International application No.

PCT/NZ01/00124

A. CLASSIFICATION OF SUBJECT MATTERInt. Cl. ⁷: C08L 77/06, 69/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: C08L 77/06, 69/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
SEE ELECTRONIC DATA BASE BELOWElectronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WPIDS, JAPIO**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 63 175 070 A (TOYOBO) 19 July 1988 (Data supplied from the esp@cenet database - 12) Abstract	1-11, 12, 13-18
X	Derwent Abstract Accession No. 89-042674/06, Class A17 (A21 A23), JP 63 314 270 A (UNITIKA) 22 December 1988 Abstract	1-11, 12, 13-18
X	Derwent Abstract Accession No. 91-112702/16, Class A23 (A13 A14), JP 03 052 952 A (DAICEL) 7 March 1991 Abstract	1-11, 12, 13-18

☒ Further documents are listed in the continuation of Box C
 ☐ See patent family annex

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"P" Document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

26 July 2001

Date of mailing of the international search report

3 August 2001

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/NZ01/00124

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 05 262 971 A (TORAY) 12 October 1993 (Data supplied from the esp@cenet database - 12) Abstract	1-11, 12, 13-18
X	Derwent Abstract Accession No. 96-236247/24, Class A13 (A12 A23 A85), JP 08 092460 A (TORAY) 9 April 1996 Abstract	1-11, 12, 13-18

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